Superconductivity and Quantum Criticality in $CeCoIn_5$

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(Dated: February 1, 2008)

Electrical resistivity measurements on a single crystal of the heavy-fermion superconductor $CeCoIn_5$ at pressures to 4.2 GPa reveal a strong crossover in transport properties near $P^* \approx 1.6$ GPa, where T_c is a maximum. The temperature-pressure phase diagram constructed from these data provides a natural connection to cuprate physics, including the possible existence of a pseudogap.

PACS numbers: 74.70.Tx, 74.62.Fj, 75.30.Mb, 75.40.-s

The relationship between unconventional superconductivity and quantum criticality is emerging as an important issue in strongly correlated materials, including cuprates, organics and heavy-fermion intermetallics [1, 2]. In each of these, either Nature or an externally imposed parameter, such as pressure or chemical substitutions, tunes some magnetic order to a $T = 0$ transition where quantum fluctuations introduce new excitations that control thermodynamic and transport properties over a broad range of temperature-parameter phase space. A frequent, but controversial [3], assumption is that these same excitations may mediate Cooper pairing in the anisotropic superconductivity that appears in the vicinity of a quantum-critical point (QCP). Indeed, the effective potential generated by proximity to a quantum-critical spin-density wave [4] also can lead [5] to anisotropic superconductivity.

Though the issue of quantum criticality and superconductivity came to prominence in the context of the cuprates (see, eg [6]), there have been several recent examples of this interplay in heavy-fermion compounds [7]. Like the cuprates, unconventional superconductivity in these Ce-based heavy-fermion materials develops out of a distinctly non-Fermi-liquid normal state that evolves in proximity to a continuous $T = 0$ antiferromagnetic transition; but unlike the cuprates, this state can be accessed cleanly by applied hydrostatic pressure and without introducing extrinsic disorder associated with chemical substitutions. In addition to chemical inhomogeneity in the cuprates, a further 'complication' is the existence of a pseudogap state above T_c for dopings less than optimal [8]. Whether evolution of the pseudogap with doping produces another QCP near the optimal T_c in the cuprates is an open question as is the origin of the pseudogap [9]. Thus far, a pseudogap has not been detected in canonical quantum-critical heavy-fermion systems, such as $CeIn_3$ or $CePd_2Si_2$ under pressure, but this may not be too surprising. In these cases, superconductivity appears only at very low temperatures, less than 0.4 K, and over a narrow window of pressures (≤ 0.8 GPa) centered around the pressure where T_N extrapolates to zero. In analogy to the cuprates, we would expect a pseudogap in these Ce compounds to exist only over a narrow temperature window, $0.1 - 0.2$ K above T_c, and in a fraction of the narrow pressure window, which certainly would make detection of a pseudogap difficult.

$CeCoIn_5$ offers the possibility of making a connection between other Ce-based heavy-fermion materials and the cuprates. Bulk, unconventional superconductivity is present in $CeCoIn_5$ at atmospheric pressure [10, 11, 12, 13] and develops out of a heavy-fermion normal state in which the resistivity is approximately linear in temperature. Application of a magnetic field sufficient to quench superconductivity reveals a specific heat diverging as $-T \ln T$ and associated low-temperature entropy consistent with its huge zero-field specific heat jump at T_c ($\Delta C/\gamma T_c = 4.5$). Together, these properties suggest that $CeCoIn_5$ may be near an antiferromagnetic quantum-critical point at $P = 0$. Its isostructural, antiferromagnetic relative $CeRhIn_5$ becomes a pressure-induced unconventional superconductor near 1.6 GPa, with a $T_c = 2.1$ K close to that of $CeCoIn_5$ at $P = 0$ [14, 15]. This and other similarities between $CeCoIn_5$ at $P = 0$ and $CeRhIn_5$ at $P = 1.6$ GPa reinforce speculation [16, 17] that the nearby antiferromagnetic QCP in $CeCoIn_5$ may be at an inaccessible slightly negative pressure. The 1.7% smaller cell volume of $CeCoIn_5$ compared to that of $CeRhIn_5$ is consistent with this view. We have studied the effect of pressure on the electrical resistivity and superconductivity of $CeCoIn_5$ and find striking correlations between them that are reminiscent of cuprate behaviors.

Four-probe AC resistivity measurements, with current flowing in the tetragonal basal plane, were made on a single crystal of $CeCoIn_5$ grown from excess In flux. Hydrostatic pressures to 5 GPa were generated in a toroidal anvil cell [18] in which a boron-epoxy gasket surrounds a teflon capsule filled with a glycerol-water mixture (3:2 volume ratio) that served as the pressure transmitting fluid. Pressure inside the capsule was determined at room temperature and at low temperatures from the pressure-dependent electrical resistivity and T_c of Pb, respectively [19]. The width of superconducting transition of Pb did not exceed 15 mK, indicating good hydrostatic conditions and providing an estimate of the pressure-measurement uncertainty, ± 0.04 GPa.

The response of $\rho(T)$ to pressure over a broad temperature scale [16, 17] is typical of many Ce-based heavy-

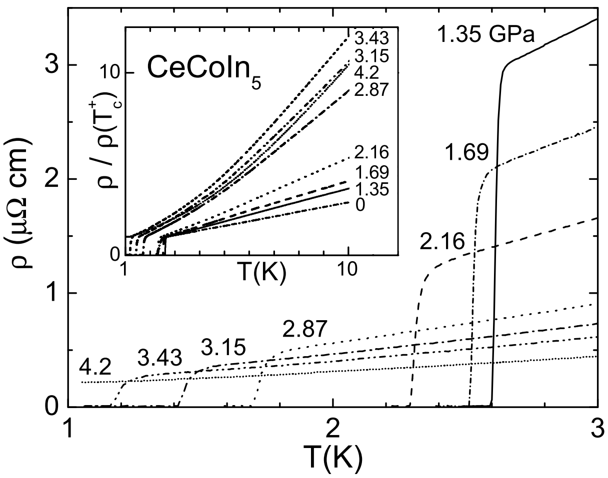

FIG. 1: Effect of pressure on the low-temperature resistivity and superconducting T_c of $CeCoIn_5$. The inset shows the resistivity ρ normalized by $\rho(T_c^+)$ up to 10 K.

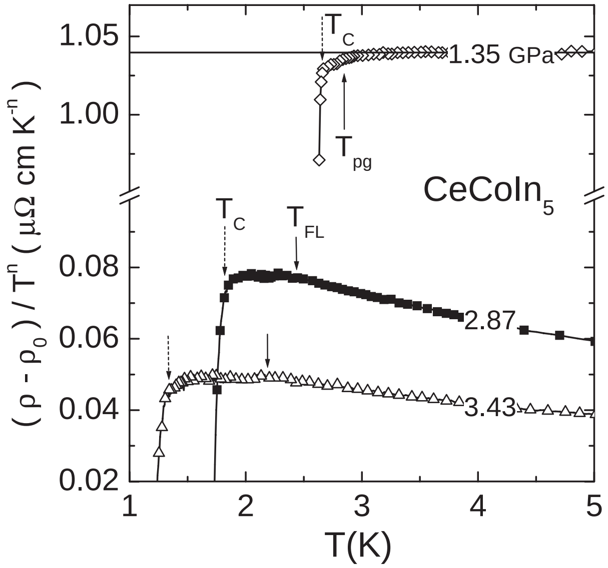

FIG. 2: Representative plots of $(\rho(T) - \rho_0)/T^n$ vs T. At low pressures, $(\rho(T) - \rho_0)/T^n$ is constant from $T_{pg} \approx (1.15 \pm 0.05)T_c$ to about 10 K. At the higher pressures, there is a well-defined range below T_{FL} where $(\rho - \rho_0) \propto T^2$ is Fermi-liquid like. The resistive onset of superconductivity, defined by the intersection of linear extrapolations of $\rho(T)$ from above and below T_c, is denoted by dashed vertical arrows. At the highest pressures, the gradual rounding just above T_c is due to a broadened transition (see Fig. 3) and not to T_{pg}

fermion compounds and reflects a systematic increase in the energy scale of spin fluctuations. Most interesting is the low-temperature response (Fig. 1) where T_c and the resistivity just above T_c ($\rho(T_c^+)$) decrease rapidly with pressures $P > 1.35$ GPa. We have analyzed the low temperature resistivity, plotted over a broader temperature scale in the inset of Fig. 1, by either fitting to a form $\rho(T) = \rho_0 + AT^n$, with ρ_0, A and n fitting parameters, or from plots of $(\rho(T) - \rho_0)/T^n$. A drawback to the former approach is that parameters can be sensitive to the temperature range chosen for fitting. The more sensitive and revealing approach is the latter in which ρ_0 and n are adjusted to give the best horizontal line. This procedure requires one less parameter, A, which is provided directly by the magnitude of $(\rho(T) - \rho_0)/T^n$. Representative examples of this approach are plotted in Fig. 2. Parameters obtained from these plots are well defined and agree, typically to 10% or better, with parameter values extracted from straightforward fits. As indicated by arrows on the lowest pressure curve in Fig. 2, $(\rho(T) - \rho_0)/T^n$ deviates from the horizontal trend at a temperature T_{pg} well above the onset of superconductivity. This departure implies either a decrease in the scattering rate or increase in carrier density. Resistivity measurements at $P = 0$ and in a magnetic field (6 T) greater than $H_{c2}(0)$ find [20] a similar departure of $(\rho(T) - \rho_0)/T^n$ from a T-independent curve at T_{pg}=3 K. Thus, T_{pg} does not originate from superconducting fluctuations or from trace amounts of free indium. We also note that this behavior was present in previous measurements on other single crystals $CeCoIn_5$ but was missed in inspections of ρ versus T curves [16]. We will return to a discussion of T_{pg} later.

Values of ρ_0, n and A obtained from these plots are summarized in Figs. 3a and b. There is an unmistakable crossover in the pressure dependence of ρ_0 and n at $P^* \approx 1.6$ GPa. Below P^*, $n = 1.0 \pm 0.1$ for $T_{pg} \leq T \leq 10$ K. This value of n is expected for a 2-dimensional, antiferromagnetic quantum-critical system [21] and is not too surprising given the layered crystal structure [22] and anisotropy in electronic states of $CeCoIn_5$ [10, 23]. At pressures greater than P^*, n rapidly approaches the Fermi-liquid value of 2.0, which holds from just above T_c to T_{FL} (see Fig. 2). Though not shown in either Fig. 2 or 3a, at these higher pressures n assumes a value of 1.5$\pm$0.1, characteristic of a 3-D antiferromagnetic QCP, in a temperature interval from slightly greater than T_{FL} to as high as $\sim$60 K. For $P \geq P^*$, ρ_0 decreases reversibly by an order of magnitude to a very small value of about 0.2 $\mu\Omega$cm. Clearly, pressure does not remove impurities from the sample; the large decrease in ρ_0 must be due to a pressure-induced change in inelastic scattering processes. Theories of electronic transport in quantum-critical systems show [24, 25] that impurity scattering can be strongly enhanced by quantum-critical fluctuations. This sensitivity provides a natural explanation for a decrease in scattering with increasing pressure, if there is a crossover near P^* from quantum-critical ($P < P^*$) to a Fermi-liquid-like state for ($P > P^*$). Such a crossover is suggested by the pressure variation of n.

Experimental values of the T^n-coefficient of resistivity, plotted in Fig. 3b, also decrease strongly with increasing pressure for $P \lesssim P^*$. For comparison, we show

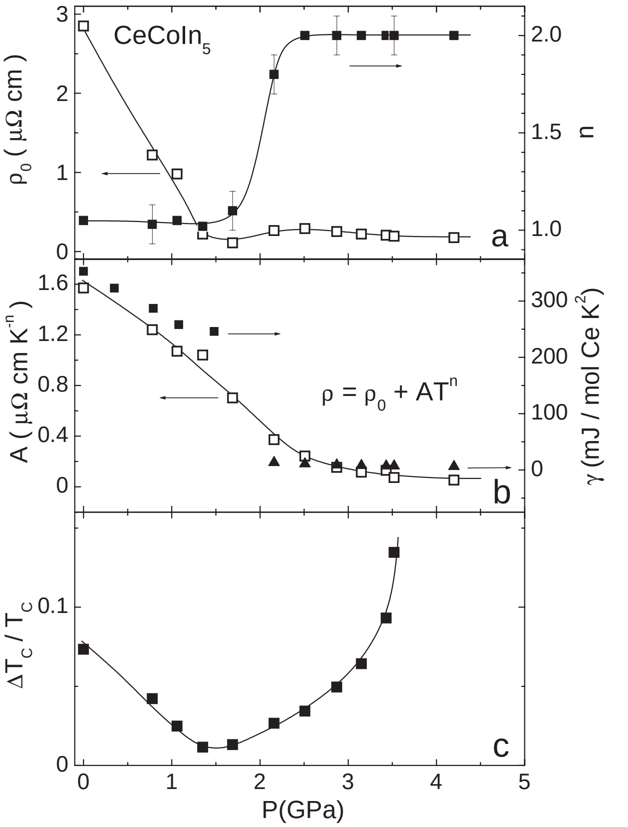

FIG. 3: (a) Values of ρ_0 and n obtained from plots as shown in Fig. 2. (b) Temperature coefficient of resistivity, A, associated with values of n given in the top panel, and specific heat coefficient γ measured directly ($P < 1.5$ GPa) and, for $P > 2$ GPa , inferred from the T^2 coefficient of $\rho(T)$. See text for details. (c) Superconducting transition width normalized by T_c. ΔT_c and T_c are the half-width of the resistive transition width and resistive midpoint, respectively. Solid lines in all cases are guides to the eye.

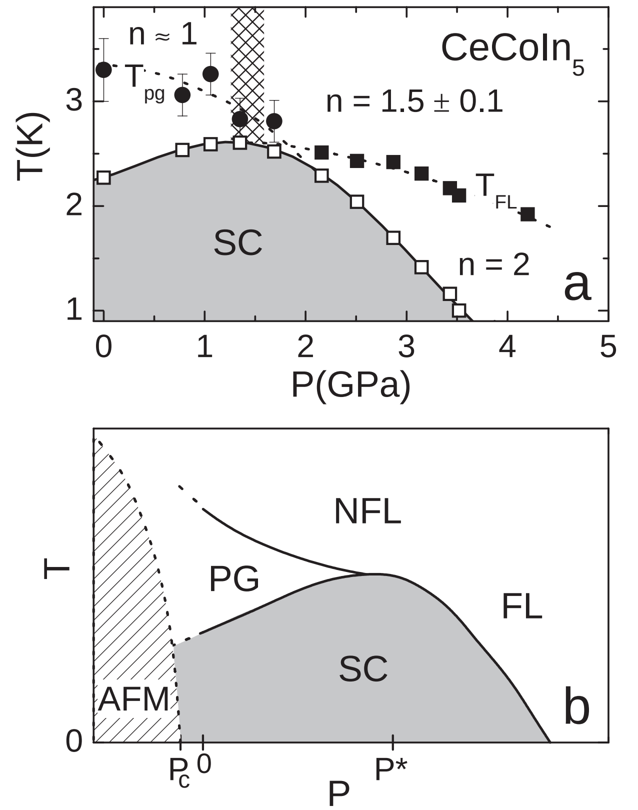

FIG. 4: (a) Temperature-pressure phase diagram for $CeCoIn_5$ constructed from data shown in Figs. 2 and 3. (b) Schematic $T-P$ phase diagram. AFM: Néel state; PG: pseudogap state; SC: unconventional superconducting state; FL: Fermi liquid; NFL: non-Fermi-liquid. See text for details.

the variation of the electronic specific heat divided by temperature ($C/T \equiv \gamma$) at 3 K for pressures less than 1.5 GPa [26] and, at higher pressures, we plot values of γ inferred from the empirical relationship $A = 1 \times 10^{-5}\gamma^2$, where A is the T^2-coefficient of resistivity, followed by several heavy-fermion compounds [27]. Though a T^2 coefficient proportional to γ^2 is expected for a Landau Fermi-liquid [6], it is not obvious *a priori* why $A(P)$ approximately tracks the directly measured γ at low pressures, but this seems to be the case. In any event, both $A(P)$ and $\gamma(P)$ exhibit qualitatively different magnitudes and functional dependencies above and below P^*. The dramatic differences in $\gamma(P)$ at low and high pressures suggest that either the empirical relationship is invalid and underestimates γ in the high-pressure regime by at least two orders of magnitude or there is a very rapid crossover in the density of low-energy excitations in the vicinity of P^*. In the Fermi-liquid-like state above P^*, we would expect the temperature scale T_{FL} (see Fig. 2) to be proportional to $A^{-0.5}$ instead of the approximate $T_{FL} \propto \sqrt{A}$ found experimentally. We do not understand this discrepancy. Direct measurements of C/T at pressures above P^* would be valuable.

Finally, we note that the resistive transition to the superconducting state is equally sensitive to the crossover at P^*. As shown in Fig. 3c, the relative transition width $\Delta T_c/T_c$ passes through a pronounced minimum near P^*. To some extent this behavior is accentuated by the pressure variation of T_c, but ΔT_c itself is a minimum near P^*. It is as if the optimally homogeneous state is singular in the vicinity of P^*.

From the data presented above, we construct a temperature-pressure phase diagram for $CeCoIn_5$ given in Fig. 4a. This phase diagram is more like that of the cuprates around optimal doping than the canonical behavior of heavy-fermion systems exemplified, for example, by $CeIn_3$ or $CePd_2Si_2$ [7]. There is no obvious long-range ordered state in $CeCoIn_5$ at atmospheric or higher pressures. Rather, in view of its relationship to $CeRhIn_5$, a reasonable speculation is that the antiferromagnetic QCP of $CeCoIn_5$ is at a slightly negative pressure. The resistivity exponent n and specific heat (ref.

[26]) are those of a non-Fermi-liquid up to P^* where T_c is a maximum. Beyond P^*, T_c drops rapidly and parameters characterizing electronic transport are qualitatively different, Fermi-liquid-like at temperatures just above T_c and those of a 3-D quantum-critical antiferromagnetic state at higher temperatures.

The generic $T-P$ phase diagram in Fig. 4b provides a broader perspective of our experimental observations and their relationship to $CeRhIn_5$. In the spirit of this diagram, the non-Fermi-liquid transport and thermodynamic properties of $CeCoIn_5$ below P^* are controlled by an antiferromagnetic QCP at the inaccessible negative pressure denoted by P_c in Fig. 4. We have chosen P_c to correspond approximately to the critical pressure at which superconductivity appears in $CeRhIn_5$. Spin-lattice relaxation measurements on $CeRhIn_5$ identify [28] a pseudogap whose signature first appears at pressures slightly less than P_c and at a temperature of $\sim$5 K and then decreases toward T_c with increasing pressure until its signature is lost at $P \gtrsim P_c$. If we associate the temperature $T_{pg}(P)$ in Fig. 4a with a resistive signature for a pseudogap and extrapolate $T_{pg}(P)$ in $CeCoIn_5$ to slightly negative pressures, there is a natural connection between our observations and properties of $CeRhIn_5$. Neither the $1/T_1T$ measurements on $CeRhIn_5$ nor our transport measurements on $CeCoIn_5$ permits definitive statements about the origin or nature of the pseudogap [29]. We note, however, that 4-fold anisotropy of the thermal conductivity in the basal plane of $CeCoIn_5$ persists [12] to 3.2 K $\approx T_{pg}$ at atmospheric pressure. If this in-plane modulation of the normal-state thermal conductivity is due to the presence of a pseudogap, it implies d-wave symmetry.

The schematic phase diagram, Fig. 4b, reflects the relationship among various phases of $CeCoIn_5$ and $CeRhIn_5$ and is similar to the T-doping phase diagram of the cuprates. This diagram and experimental data from which it is inferred indicate that the physics of heavy-fermion systems may be more closely related to that of the cuprates than previously appreciated. Aside from the structural layering in $CeCoIn_5$ and $CeRhIn_5$ and their relatively high T_c's, they are similar in many respects to other Ce-based heavy-fermion materials in which superconductivity develops near a QCP. Further study of $CeCoIn_5$, in parallel with the cuprates and other pressure-induced heavy-fermion superconductors, holds promise for bridging our understanding of the interrelationship between unconventional superconductivity and quantum criticality across these interesting classes of correlated matter.

We thank R. Movshovich for helpful discussions. VS acknowledges S. M. Stishov for his support during the initial stage of this research. Work at Los Alamos was performed under the auspices of the US Department of Energy.

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